Scaling exponent within the side-jump mechanism of Hall effect size-dependence in Ni nanocrystals

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High-resolution Hall data in only 3.5 μg of Ni nanocrystals, grown in a planar array on TiN, are reported. We conclude from the exponent, $n \sim 1.06 \pm 0.01$ in $R_s \sim \rho^n$, where R_s is the extraordinary Hall constant and ρ is the Ohmic resistivity, that the side-jump mechanism could still be operative if the nanocrystals are below a certain critical size and the mean free path of the electrons is strongly temperature dependent only in the magnetic layer. Also, the 1000 times larger value of R_s than those in bulk Ni makes it an ideal candidate for magnetic sensors. © *2008 American Institute of Physics*.
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The Hall effect in semiconductors is the basis of many devices in measuring magnetic fields. In nonmagnetic metals, the ordinary Hall coefficient is low because of the high carrier density. The interest in the Hall effect has recently increased because its value for ultrathin single ferromagnetic metal (e.g., Ni, Fe) or bimetal (e.g., $Fe_{0.5}Pt_{0.5}$, $Co_{0.9}Fe_{0.1}/Pt$) films[1] is found to be significantly greater than those of the bulk and is often comparable to the best values in semiconductors. The growing interest in the extraordinary Hall effect (EHE) also lies in its use as a simple and efficient tool in magnetic characterization of thin films and nanometer size magnetic entities. When we reduce the size of the magnetic particles to a nanometer or so, several interesting properties show up, such as giant magnetoresistance and superparamagnetism, among others.[2,3] The origin of the spontaneous/EHE and its scaling with the Ohmic resistivity is very well understood in *bulk homogeneous* ferromagnets where the Hall resistivity ρ_H is given by[4]

$$\rho_H = R_0 B + \mu_0 R_s M, \tag{1}$$

where B is the magnetic induction, M is the magnetization, and R_o and R_s are the ordinary and the extraordinary Hall constants, respectively. The first term represents the ordinary Hall effect while the second term, coming from the extraordinary/spontaneous Hall effect, is a characteristic of ferromagnetic materials, and is proportional to its magnetization. The origin of the EHE lies in the spin-orbit interaction present in a ferromagnet. R_s obeys a power law relationship with the electrical resistivity ρ, given by $R_s = \alpha\rho^n$, where α is a constant. Smit's classical asymmetric scattering[5] gives the exponent $n=1$ while the quantum mechanical side-jump scattering[6] yields $n=2$.

Epitaxially grown ferromagnetic 50 nm thin films[7] of $Sr_{1-x}Ca_xRuO_3$, with Curie temperature and magnetization (M) disappearing at $x \sim 0.7$, show that the Hall conductivity scales with $M(T)$. A work, more related to ours and recently reported[8] in 2008 in 5–150 nm thin films of magnetite, shows that in the dirty regime ($\rho \gg 100 \times 10^{-8}$ Ω m), $n \sim 1.6$ over 4 decades of ρ, independent of temperature and film thickness. In layered ferromagnetic[9] Fe dichalcogenide TaS_2 of thickness ~ 20 μm (bulk), the inelastic part of the Hall conductivity is found to scale with ρ^2. In this letter, we explore this scaling aspect of the Hall effect in epitaxially grown *single* layer of Ni nanoparticles sandwiched between conducting TiN on a sapphire substrate. We conclude that the scaling exponent n depends on the size of the magnetic nanoparticles as predicted by the theory developed by Zhang[10] for magnetic/nonmagnetic multilayers. This work presents a major advance in the analysis and interpretation of the extraordinary Hall data for inhomogeneous ferromagnets.

A multitarget pulsed laser deposition (PLD) system was used for sample preparation where two polycrystalline Ni and TiN targets were alternately ablated onto (1000) single crystal sapphire substrate glued to a resistive heater at 600 °C with the chamber at 10^{-7} Torr. The details are reported in our previous publication.[11] The shape and size of Ni nanoparticles and the thickness of the TiN layer were investigated by cross-sectional scanning transmission electron microscopy (STEM) as shown in Fig. 1. It is evident that the bottom TiN, which serves as a diffusion barrier buffer layer between Ni particles and sapphire, is significantly thicker (30 nm) than the top TiN layer (15 nm) serving as a protective passive layer for highly reactive Ni nanoparticles from the air ambient. The growth of Ni nanoparticles takes place via Volmer–Weber island growth mode on a TiN buffer layer. It should be noted that the epitaxial quality of the TiN buffer layer on sapphire is extremely high with well defined in-plane orientation relationship of $(111)_{TiN} \| (0001)_{sapphire}$ and $[2\bar{2}0]_{TiN} \| [30\bar{3}0]_{sapphire}$. Besides, the Ni nanoparticles are well oriented on the TiN layer, particularly establishing local epitaxy at their interfaces. The major orientation relationship between Ni and TiN on the sapphire substrate is described as $(111)_{Ni} \| (111)_{TiN}$ and $[2\bar{2}0]_{sapphire} \| [2\bar{2}0]_{TiN}$. The Ni nanoparticles are grown in a truncated pyramidal-shaped slab of average base length of ~ 120 nm and height of ~ 25 nm and are bounded by wide top and bottom (111) planes on TiN-buffered sapphire. As

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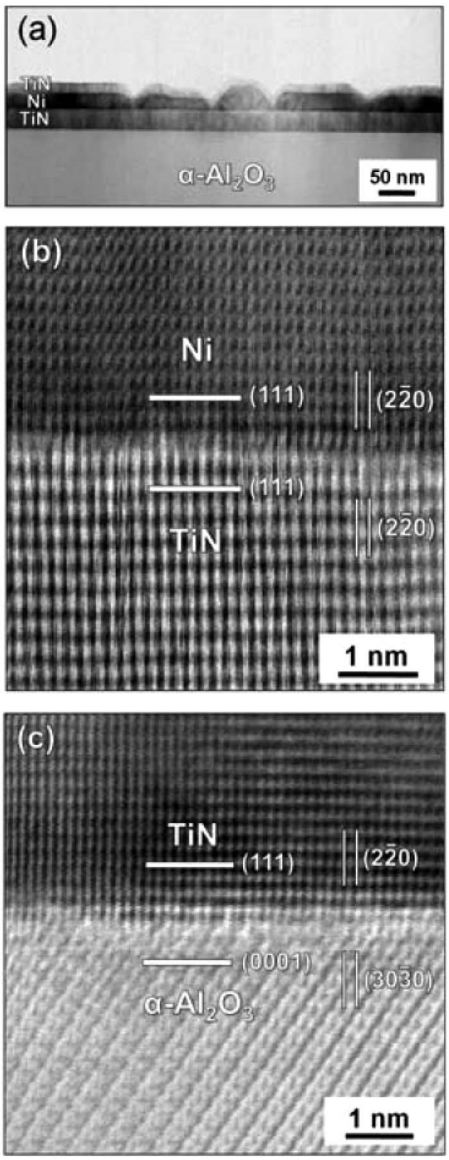

FIG. 1. STEM images of the Ni particles deposited on TiN-buffered sapphire substrate. (a) Low-magnification overview image of Ni particles, (b) high-magnification image showing the interface between Ni and TiN buffer layer, and (c) high-magnification image of the TiN/Sapphire interface.

seen in Fig. 1(b), the interface between the Ni nanoparticle and the surrounding TiN is atomically sharp, indicating the presence of pure metallic Ni at the surface. Our previous atomic level structural characterization study, through STEM and electron energy loss spectroscopy on nanoparticles of Ni embedded in Al_2O_3 thin film matrix using PLD and similar deposition conditions, proved the absence of any oxide layer on the Ni nanoparticle surface.[11] Thus, based on an atomically sharp interface between Ni–TiN, it is reasonable to assume that the Ni nanoparticles in the TiN matrix are very unlikely to have any appreciable nitride layer that could be magnetically dead.

The magnetization versus temperature and field $M(T,H)$ measurements were carried out using a vibrating sample magnetometer interfaced with a seven-tesla physical property measurement system (PPMS) of Quantum Design. All the transport measurements that include resistance versus temperature and Hall measurements were also carried out in PPMS using five-wire ac transport option. At every temperature and in zero fields, the Ohmic voltage, due to the misalignment of the Hall probes, was very carefully compensated to the level of the noise of the circuit. Even after balancing the zero-field offset, we performed the Hall measurements at both positive and negative fields so that the magnetoresistive component can be subtracted out from the measured Hall resistivity.

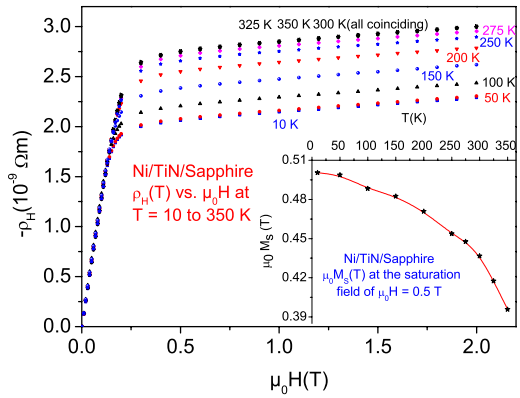

FIG. 2. (Color online) Hall resistivity $(-\rho_H)$ is plotted against external magnetic field $(\mu_0 H)$ at several temperatures from 10 to 350 K. The inset shows magnetization $\mu_0 M_S$ vs temperature data at the saturation field $\mu_0 H$ of 0.5 T.

Figure 2 is the plot of the Hall resistivity $(-\rho_H)$ versus the applied field $(\mu_0 H)$, measured at different temperatures. The resolution of the data is excellent considering the fact that the actual mass of Ni, which produces the EHE, is only ~ 3.5 μg $(4.4\times 10^{-7}$ cc). The Hall coefficients R_0 and $\mu_0 R_s M_S$ were found as the slope and the intercept of the linear portion of the Hall curves above the saturation field. The $\mu_0 M_S(T)$ data at the saturation field $\mu_0 H = 0.5$ T, after diamagnetic correction from the sapphire substrate, are shown in the inset of Fig. 2. The curve is convex upwards decreasing faster than linear with temperature proving the ferromagnetic state of Ni makes the observation of the spontaneous Hall effect possible. $\mu_0 M_S(T)$ was then substituted at each temperature in $\mu_0 R_S M_S$. Here, both R_0 and R_S are found to be negative. At 300 K, the modulus of R_s (since it is negative) is about 11 times larger than that of bulk Ni, while at 10 K, it is nearly three orders of magnitude larger. The resistivity of TiN is weakly temperature dependent between 10 and 350 K (typically $\sim 45\times 10^{-8}$ Ω m at 300 K) and is markedly different from that of nanocrystalline Ni (typically 0.55×10^{-8} Ω m at 10 K and 7.5×10^{-8} Ω m at 300 K for 100 nm size). Our measurements (not shown) on the temperature dependence of the resistivities of the composite Ni/TiN/Sapphire sample and the nanocrystalline 100 nm Ni samples show that at lower temperatures, the individual resistivities are widely different and hence, the current density through the Ni content is actually much larger than the measured current. This fact is indeed the reason behind the much larger value of the extraordinary Hall resistivity and hence the apparent value of R_s. Following our earlier work[12] on a sample of five bilayers of *isolated* Ni nanoparticles and TiN on silicon substrate, we calculate the current density J_{Ni} that passes through the nickel and correct the Hall resistivity, now given by $\rho_H = E_y / J_{\mathrm{Ni}}$.

In Fig. 3, we plot ln R_s (with corrected R_s) versus ln ρ (ρ being the resistivity of the nanocrystalline Ni). The scaling exponent $n = 1.06 \pm 0.01$ is obtained from the excellent linear fit from 10 to 350 K with a correlation coefficient of

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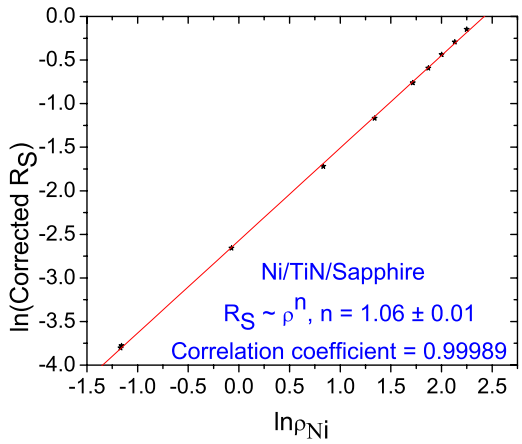

FIG. 3. (Color online) ln R_s (corrected) is plotted against ln ρ_{Ni} (resistivity of 100 nm nanocrystalline Ni) from 10 to 350 K. The scaling exponent n is found to be 1.06 ± 0.01.

0.999 89. The average thickness of Ni in this sample is ~25 nm and its resistivity is $(70-93)\times 10^{-8}$ Ω m (4.2 to 300 K). In contrast, the sample of our earlier work[12] had a higher thickness of Ni at $\sim 5\times 13 = 65$ nm, a lower corresponding resistivity of $(47-54)\times 10^{-8}$ Ω m, and $n \sim 1.7$, nearly the same as in bulk Ni ($n=1.68$). We explain this thickness dependence of n in terms of the theory of Zhang[10] for magnetic/nonmagnetic multilayers. To derive the Hall resistivity, ρ_{yx} from the side-jump mechanism, impurity, and interface scattering are represented by a scattering potential that includes the spin-orbit coupling (λ_{SO}) between the conduction electrons and the impurities. The spin-orbit scattering produces the Hall resistivity through the side-jump and the skew scattering. Since the resistivities of the multilayer samples are large compared to those of the bulk, the side-jump contributes most to the EHE. For *homogeneous magnetic materials*, the Hall resistivity, ρ_{yx}, and hence R_S is simply proportional to ρ^2. However, for the present structure, the Hall conductivity σ_{yx} can be obtained in two limiting cases where the mean free path (MFP) is $\ll$ the layer thickness and the scaling law again becomes valid and in the long MFP limit (Co–Cu, Fe–Cr, etc.) where the MFP $\gg$ the layer thickness and hence the electrons pass through many layers before getting scattered. The Hall conductivity σ_{yx} now depends on the ratio of relaxation times in magnetic and nonmagnetic layers. In this limit, $\rho_{yx} \sim \rho^2$ is no longer valid as found in various experiments giving "n" anywhere between ~1 to ~4. The above theory finally shows that $n<2$, $n=2$, or $n>2$ when the MFP of only the magnetic layer, or the nonmagnetic layer, or both the layers, but in a fixed ratio, respectively, is temperature dependent. The calculated Hall resistivity versus layer thickness (0.5 to 50 nm), *assuming* some fixed MFPs, shows that the ordinary scaling law with $n=2$ is valid for a layer thickness of more than 10 nm. This limit will certainly change from one system to the other since the scattering parameters could be altogether different.[10]

The thickness dependence of n, found by us, is thus very consistent with the predictions of the above theory, especially the fact that samples with a thickness greater than some critical value, depending on the scattering parameters, could give $n \sim 2$. We have indeed found that the resistivity is strongly temperature dependent in the magnetic layer (Ni) and very weakly in the nonmagnetic layer (TiN), exactly the condition which could yield values of n much lower than 2, as in our single magnetic Ni layer (~25 nm Ni) even when side-jump is *the* scattering mechanism. However, a much higher thickness of Ni (65 nm), such as that of the five-bilayer Ni/TiN sample, could again yield $n \sim 2$. We must emphasize that the closeness of n to the value "1" might be erroneously interpreted as due to the predominance of Smit asymmetric scattering over the side-jump mechanism. Skew scattering cannot dominate until such high temperatures of 350 K.

A similar work[1] on planar arrays of Fe nanoparticles embedded in Pt thin film and another on a planar array of Co nanoparticles in the Pt matrix[13] had reported EHE in the ferromagnetic and superparamagnetic phases, respectively. However, none[1,13] had given any analysis for the scaling exponent appropriate for such composite nanosystems. A much older work[14] on metal-insulator film $(Ni)_x-(SiO_2)_{1-x}$ of varying metallic Ni had shown the so-called "giant hall effect" where the Hall resistivity ρ_H increased by four orders of magnitude for $x \sim 0.55$, the percolation threshold. These materials thus become good candidates for high-sensitivity Hall sensors. The scaling exponent n was found to be ~0.7 but *no* explanation was offered for this low value.

To conclude, the key results of this Hall effect studies in Ni/TiN/Sapphire are: (a) The scaling exponent depends on the average size of the magnetic nanoparticles, e.g., $n \sim 1$ for the present 25 nm Ni sample as against ~1.7 (bulk value) for a much thicker 65 nm Ni sample of our earlier studies[12] and (b) three orders of magnitude larger value of the extraordinary Hall constant, R_S here than that in bulk Ni makes it an ideal candidate for good magnetic sensors.

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